

10/523,373



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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Joachim Kiefer, Oemer Uensal and Gordon Calundann

Application No.: 10/523,373 Group: 1713

Filed March 23, 2005 Examiner: Michael Bernshteyn

Confirmation No.: 3591

For: PROTON-CONDUCTING POLYMER MEMBRANE COMPRISING A  
POLYMER WITH SULPHONIC ACID GROUPS AND USE THEREOF IN  
FUEL CELLS

CERTIFICATE OF MAILING OR TRANSMISSION	
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DECLARATION THOMAS J. SCHMIDT UNDER 37 C.F.R. §1.132

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Dr. Thomas J. Schmidt am a citizen of the Federal Republic of Germany and reside at Frankfurt, Germany, hereby declare and say as follows:

1. I am a fully trained chemist, having studied and graduated in chemistry at University of Ulm, Germany and prepared my ph.D. thesis at University of Ulm in Fuel Cell Research. I held positions at Ernest Orlando Lawrence Berkeley National Laboratory in Berkeley, CA and at Paul-Scherrer-Institute, Switzerland, both in applied fuel cell research and development.

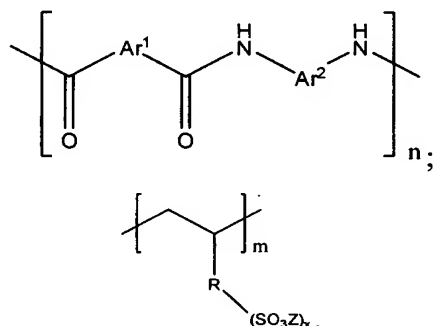
I am well acquainted with technical English.

2. I, Dr. Thomas J. Schmidt state that I have more than 5 years scientific practice in applied fuel cell membrane and membrane electrode assembly work and that I consider myself qualified by my education, knowledge and many years of experience to make this Declaration. I currently hold the position of R&D Director at BASF Fuel Cell GmbH (previously named PEMEAS GmbH).
3. . I have thoroughly read the application U.S. Serial No. 10/523,373, the office actions issued by the United States Patent & Trademark Office (hereinafter the "USPTO") for this application, the references cited by the Examiner in the office actions and the responses filed on behalf of Pemeas GmbH.
4. The claimed invention is a proton-conducting polymer material that is used as a membrane or as a coating on, for example, an electrode. The proton-conducting material of the present invention is prepared by the process recited in Claims 25 and 44. This process comprises preparation of two distinct polymers: a polyazole polymer and a polyvinyl polymer. The polyvinyl polymer includes sulfonic and/or phosphonic acid moieties. The resulting material is, therefore, a mixture of two different polymers.
5. The process for preparation of proton conducting membranes and coating of the present invention begins with mixing the starting materials for the two subsequent polymerization reactions. The starting materials for the polyazole polymer are aromatic or heteroaromatic compounds having one or more amino groups and one or more carboxylic acid groups. Examples of such compounds are listed on pages 13-15 of the English translation of the present application. The starting materials

for the polyvinyl polymer are vinyl-containing sulfonic and/or vinyl-containing phosphonic acids.

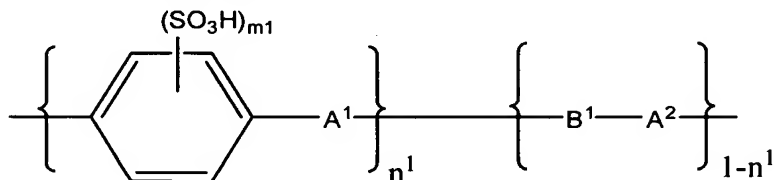
6. Following the step of mixing, the polyazole polymer is prepared by a condensation reaction between amino substituents and carboxylic substituents on the aromatic or heteroaromatic compounds. This reaction is described on page 26, lines 22-29 of the English translation of the present application. This reaction is performed by heating the mixture described above up to 350 °C and is accompanied by formation of water, which is removed by distillation or by a drying agent. Examples of resulting polymers are listed on pages 16-19 and pages 22-26 of the English translation of the present application. The vinyl-containing component of the original mixture remains unaffected by this reaction.
7. Following the formation of polyazole polymer, the resulting material is formed into a desired shape, such as a sheet. For example, the resulting material can be applied to a support, such as an electrode.
8. The concluding step in the process by which proton-conducting membranes and coating of the present invention are formed is the step in which the polyvinyl polymer is formed. This step is described on page 31, lines 10-21, and on page 32, line 16 through page 33, line 5 of the English translation of the present application. Polymerization of vinyl-containing sulfonic or phosphonic acid is a free-radical polymerization process which may be facilitated by addition of a free-radical initiator. The initiator can be added into or sprayed onto the material formed after completion of the polyazole condensation reaction (see page 31, line 23 through page 32). The polyazole component of the proton-conducting material, formed at a previous step by condensation reaction, remains unaffected by the free-radical polymerization of vinyl-containing monomers.

9. The proton-conducting material membranes or coating produced by the process described above comprises two different polymeric materials: polyazole and polyvinyl. The latter includes units substituted with sulfonic and/or phosphonic acid moieties. The two polymeric materials for a physical mixture, but are not chemically bonded. Non-limiting examples of the two polymers comprising the proton-conducting membranes are shown below:

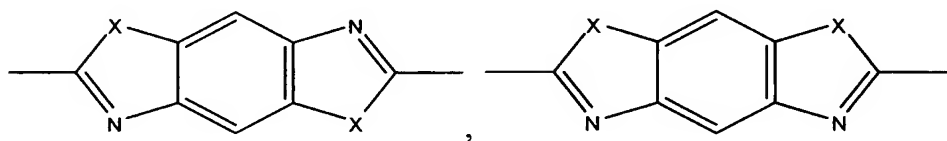


- In the above-reproduced formulas,  $\text{Ar}^1$  and  $\text{Ar}^2$  are each independently aromatic tetraamino compounds substituted with one or more aromatic carboxylic acids (or carboxylic acid derivatives such as esters, acid halides, or acid anhydrides); or aromatic or heteroaromatic diaminocarboxylic acids. R is a bond, a C1-C15 alkyl group, C1-C15 alkoxy group, ethyleneoxy group or C5-C20 aryl or heteroaryl group, wherein the above radicals are optionally substituted in turn by halogen, -OH, COOZ, -CN,  $\text{NZ}_2$ . Z is hydrogen, C1-C15 alkyl group, C1-C15 alkoxy group, ethyleneoxy group or C5-C20 aryl or heteroaryl group, wherein the above radicals are optionally substituted in turn by halogen, -OH, -CN.
10. The Examiner cited US2004/0062969 by Sakaguchi *et al.* ("Sakaguchi"). I have thoroughly studied the cited reference, and assert that the polymer disclosed by Sakaguchi is physically and chemically different from the proton-conducting material of the present invention.
11. Sakaguchi teaches a polymer compound for use as a solid polymer electrolyte membrane. The polymer of Sakaguchi is a polybenzazole compound having

sulfonic acid and/or phosphonic acid groups. This compound is represented by formulas (1) – (2), reproduced below:



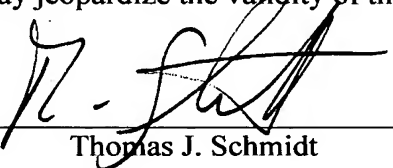
The definitions of variables  $\text{A}^1$ ,  $\text{A}^2$  and  $\text{B}^1$  are provided in paragraphs [0127] through [0129] of Sakaguchi. From those definitions it follows that variables  $\text{A}^1$  and  $\text{A}^2$  may each, independently, represent moieties shown below:



and variable  $\text{B}^1$  represents an aromatic moiety. Thus, the compound of Sakaguchi is different from the proton-conducting material of the present invention in at least two aspects. First, the compound of Sakaguchi is a single polymer as opposed to a physical mixture of two polymers. Second, the compound of Sakaguchi does not have vinyl-containing units substituted with sulfonic and/or phosphonic acid moieties. Rather, the acidic functionalities of Sakaguchi are attached to an aromatic moiety.

12. It follows from Sections 5-11 of the present Declaration, that the proton-conducting material of the present invention is both physically and chemically distinct from the polymer disclosed by Sakaguchi. First, the material of the present invention is a physical mixture of two polymers, rather than a single polymer, as disclosed by Sakaguchi. Second, one of the two polymers that comprise the proton-conducting material of the present invention is a polyvinyl substituted with an sulfonic and/or phosphonic acid moiety. The polymer of Sakaguchi does not comprise vinyl units; sulfonic and/or phosphonic functionalities are attached to an aromatic moiety.

13. I further declare that all statements herein of my own knowledge are true and that all statements made on information in belief are believed to be true; and further that the statements are made with the knowledge that willful false statements of the like so made are punishable by fine or imprisonment or both Under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereof.

  
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Thomas J. Schmidt

24-05-2007  
Date